

Particle Emission Measurements from L-Category Vehicles

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ABSTRACT

In 2011 a particle number (PN) limit was introduced in the European Union's vehicle exhaust legislation for diesel passenger cars. The PN method requires measurement of solid particles (i.e. those that do not evaporate at 350 °C) with diameters above 23 nm. In 2013 the same approach was introduced for heavy duty engines and in 2014 for gasoline direct injection vehicles. This decision was based on a long evaluation that concluded that there is no significant sub-23 nm fraction for these technologies. In this paper we examine the suitability of the current PN method for L-category vehicles (two- or three-wheel vehicles and quadri-cycles). Emission levels of 5 mopeds, 9 motorcycles, 2 tricycles (one of them diesel) and 1 quad are presented. Special attention is given to sub-23 nm emission levels. The investigation was conducted with PN legislation compliant systems with particle counters measuring above 23 nm and 10 nm. In addition other approaches using catalytic strippers and counters from 3 nm or particle sizers were used to confirm the nature of the particles. The results showed that there is a significant portion of solid particles not counted with the current PN method. On the other hand, the high amount of volatile material can lead to artifacts below 23 nm, and thus great care has to be taken when measuring the PN of this category.

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INTRODUCTION

Mopeds (combustion engine with displacement <50 cm³ and speed <45 km/h) and motorcycles (displacement >50 cm³, speed >45 km/h) are widespread means of transport in some regions such as southern Europe and in countries of East and South Asia. In Europe (EU27) more than 37 million were registered until 2013 [1], and according to the latest data available, in 2014 more than 1 million were registered in the EU. Italy is the leading manufacturer of motorized 2-wheel vehicles in Europe. The production in 2011 was 414.000 vehicles, more than half of the European output. The country has the largest number of powered two-wheelers on the road in Europe: 8.6 million vehicles in 2011 (2.2 million mopeds and 6.4 million motorcycles) [2].

Urban air pollution can be strongly influenced by mopeds and motorcycles' emissions, which are not negligible when compared with those of modern passenger cars [3, 4]. Two stroke mopeds emit significant amounts of primary organic aerosol (POA), aromatic volatile organic compounds (VOC) and also produce significant secondary organic aerosol (SOA) [5, 6]. The relative contribution of mopeds and motorcycles to particulate matter (PM) emissions started to increase after 2011. The reason is the introduction of diesel particulate filters (DPF) in both passenger cars and heavy duty vehicles at Euro 5/V and Euro 6/VI levels, which significantly decreased emissions from these vehicle categories. Although the contribution of mopeds and motorcycles to total road transport PM emissions will probably remain small (~5%) by 2020, their contribution could increase to ~20% of when focusing on urban

emissions [7]. These values could be even higher when particle number (PN) is considered because of the high number of nuclei particles formed from the unburnt fuel and lubricant.

PN measurements of mopeds and motorcycles have been conducted by some researchers (e.g. [8, 9, 10, 11, 12]) but for the latest Euro 3 the data are limited (e.g. [10]). The emission levels are usually much higher than the current emission limits for passenger cars (6×10^{11} p/km for solid particles >23 nm), in some cases at levels close to diesel vehicles without DPF [3, 4]. Sub-23 nm solid PN measurements are rare [13]. It has been mentioned that the PN measurements of mopeds are prone to artifacts due to the high concentration of volatile materials [14, 15].

Regulation (EU) No 168/2013 sets out environmental requirements for two stages with the second stage (Euro 5) being mandatory for new types of vehicles as of 01 January 2020. An Environmental Effect Study stipulated in its Article 23(4) and 23(5) will provide additional underpinning through experimental testing, modelling, technical feasibility and cost-effectiveness analysis based on the latest available data. In addition, this study will assess (see Recital 12) the feasibility of in-service conformity testing requirements, off-cycle (i.e. real driving) emission requirements and a particulate number emission limit for certain (sub-) categories.

Objective of this study is to give the PN emission levels of various L-category vehicles (two- or three-wheel vehicles and quadri-cycles) following the legislation applied to passenger cars. The geometric mean diameter of the emitted size distribution will also be given.

However, due to the particular nature of their emissions (i.e. high percentage of unburnt fuel and lubricant) the sampling protocol and the sampling location will be investigated. Topics like appropriate dilution and thermal pre-treatment and their effect on the results are discussed, especially for sub-23 nm measurements. Finally, a correlation between the PN emissions of the current type approval cycles with the future world harmonized motorcycle testing cycles (WMTC) is conducted for the first time according to our knowledge. The present study is not an output of the above mentioned Effect Study, but a preliminary feasibility study that may serve as an input to the Effect Study.

EXPERIMENTAL

Set-up

The tests were conducted in the Vehicle Emissions Laboratory (VELA 1) of the Joint Research Centre (JRC) in Ispra, Italy. VELA 1 is a climatic emission test cell with a single axis roller dynamometer used mainly for motorcycles. The exhaust of the vehicles was connected to a full dilution tunnel with constant volume sampling (CVS) (Figure 1). Typical flow rates that were used in the dilution tunnel were 4-6 m³/min. In the case of 2-stroke mopeds, where high amounts of unburnt fuel and lubricant are expected, a cyclone was used at the exhaust line to remove big droplets which can harm the instrumentation, without affecting the number of fine particles. The diameter with 50% penetration, which depends on the exhaust flow rate of the vehicles, was >10 µm. The exhaust flow rates of the 50 cm³ mopeds were around 0.1-0.25 m³/min, and >1 m³/min at high speeds for the big motorcycles. They were estimated by the difference of the total CVS flow rate minus the dilution air entering the tunnel. The uncertainty of determining a small flow from two big flows may be high. For engines of 400 cm³ we compared the estimated flow with an exhaust flow meter (Sensors, 1.5 inches) and we found a difference of only 10%.

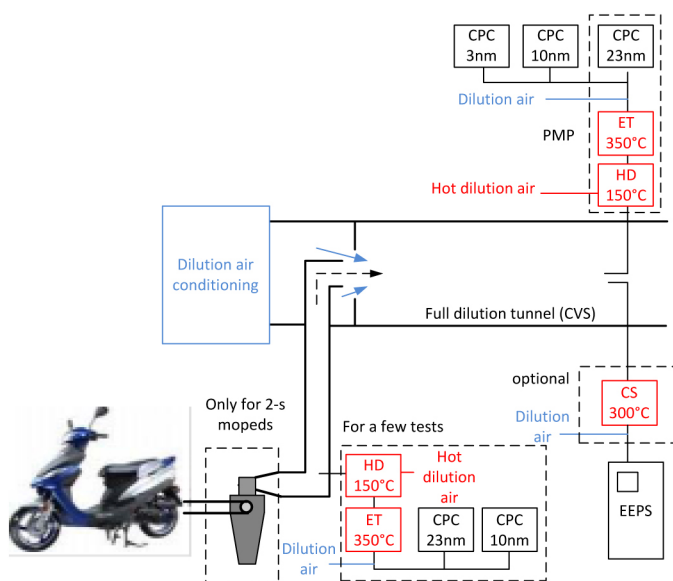


Figure 1. Experimental set-up.

From the dilution tunnel an Engine Exhaust Particle Sizer (EEPS) (TSI, 3090) was measuring the size distributions (5.6-560 nm) in real time [16]. A catalytic stripper (CS) at 300 °C (prototype from AVL) [17] was used in half of the tests (see Table 1) to remove the volatiles and measure only the non-volatiles (solids, from now on) size distribution. A short heated tube at 300 °C was included in the device upstream of the CS (residence time approximately 0.1 s). Dilution air was added at the exit of the CS to reach the flow required by the EEPS. This mixing resulted in a dilution of 10:1, approximately. The losses of the CS are 10% at sizes >30 nm, around 40% at 15 nm and 60% at 7 nm. The EEPS was compared with a Scanning Mobility Particle Sizer (SMPS) (model TSI 3080L with a TSI 3025 Condensation Particle Counter (CPC)) measuring soot particles from a propane diffusion flame generator (APG from AVL). The Geometric Mean Diameters (GMD) were within 5% for (EEPS) GMDs between 25 and 40 nm. For smaller or bigger sizes for every 10 nm the EEPS diameter should be increased 20%. The sizes presented in this paper are the EEPS sizes without any size correction. Recently TSI released an inversion algorithm that takes into account the different charging probabilities of fractal particles. We compared our corrections with the new algorithm and we found very good agreement (within 5-10 nm).

A PN measurement system compliant with the light duty vehicles (UN-ECE Reg. 83) and heavy duty engines (UN-ECE Reg. 49) regulations was used (AVL APC 489) at the dilution tunnel [18]. It consists of a hot dilution (HD) at 150 °C and an evaporation tube (ET) at 350 °C. The system was calibrated by the manufacturer and the dilution including the particle losses (as average of 30, 50 and 100 nm) is called Particle number Concentration Reduction Factor (PCRF). Typically, a primary PCRF of 1000 and a secondary of 10 were used for the mopeds and 100×10 for the rest vehicles. However, different settings were also investigated. We will use the term 'Hot dilution with evaporation tube' (HD+ET) to characterize the specific thermal pre-treatment system. The extra losses at 15 nm of this system are approximately +40%.

Downstream of the thermal pre-treatment system a TSI CPC 3790 with 50% counting efficiency at 23 nm ($d_{50\%}=23$ nm) was measuring. In parallel a TSI CPC 3772 ($d_{50\%}=10$ nm) and TSI CPC 3025A ($d_{50\%}=3$ nm) were used in order to investigate the presence of particles <23 nm. Note that HD+ET with a CPC $d_{50\%}=23$ nm is the protocol defined in the Particle Measurement Program (PMP) and used for the regulatory measurements [19].

For a few tests a PMP system was connected to the tailpipe to investigate the differences between the different sampling locations. A PCRF of 125×5 was used. The extracted flow was approximately 1.5 lpm, thus had only a negligible effect on the CVS results.

Vehicles

The vehicles are usually classified as mopeds, motorcycles and tricycles. With Directive 2002/24/EC and later Regulation (EU) No 168/2013 the vehicles are part of L-category. Details can be found in the Appendix (Table A2). In this paper the vehicles that were tested were classified as (see also Table A4 in the Appendix):

- L1e-B (2s) (Moped, <50 cm³, 2-stroke)

- L1e-B (4s) (Moped, <50 cm³, 4-stroke)
- L3e-A1 and A2 (Motorcycle, <140 km/h, 4-stroke)
- L3e-A3 (Motorcycle, >140 km/h, 4-stroke)
- L5e-A (Tricycle)
- L5e-B (Commercial tricycle, diesel)
- L7e-A1 (Quad)

The vehicles tested spanned from Euro 1 to Euro 3.

Test Cycles

The vehicles were tested according to their type approval driving cycle (UN-ECE Reg. 40, UN-ECE Reg. 47) (from now R40 and R47), but also according to the future World Harmonized Motorcycle Test Cycle (WMTC) depending on their class (details in [Table A4](#) in the Appendix and references therein). PN measurements are in real time, thus it was possible to divide the emissions in parts of the cycles (e.g. cold start part, hot part and high speed part), even if the cold part was not required by the legislation for some of them. This way the emissions from the previous cycles can be correlated to the future WMTC results. Details of the cycles and their cold, hot and high speed parts can be found in the Appendix, [Figures A1, A2, A3](#).

Some motorcycles were tested at constant speeds in order to investigate the effectiveness and robustness of the PN measurements. For these tests some parameters of the PN instruments were changed (e.g. PCRF).

Reference fuel with 5% ethanol content was used. The lubricant used was semi-synthetic.

Test Protocol

No pre-test condition is required for the R40 and R47 cycles, however all motorcycles were preconditioned by running the legislation cycle the day before to guarantee reproducible conditions. Prior to WMTC tests, all motorcycles were pre-conditioned with the WMTC the day before as stated in the regulation. At least two repetitions were conducted for each vehicle for the type approval cycle and the appropriate WMTC ([Table 1](#)). Although error bars will not be given for all cases the difference between two identical cycles were typically 10-15% for solid particles and 20-25% for total particles. [Table A4](#) in the Appendix summarizes the tested vehicles, the relevant type approval cycles, their class and the future type approval WMTC.

RESULTS AND DISCUSSION

Initially, the exploratory results regarding the sampling location and sampling conditions will be given. Then, the measured emission levels will be presented. Finally, the correlation of the various cycles will be shown.

Table 1. Test protocol. Numbers are the repetitions conducted.

Code	R40 & R47	WMTC	Extra investigations
L1e-B (2s) #1	2	-	-
L1e-B (2s) #2	6	4	Repeatability, thermal treatment
L1e-B (4s) #3	6	1	Tailpipe
L1e-B (4s) #4	4	2	
L1e-B (4s) #5	3	3	Tailpipe
L3e-A1 #1	2	2	Tailpipe
L3e-A2 #2	4	4	
L3e-A2 #3	2	3	Thermal treatment
L3e-A2 #4	1	1	
L3e-A2 #5	2	3	
L3e-A2 #6	3	2	
L3e-A3 #7	2	2	
L3e-A3 #8	-	4	
L3e-A3 #9	-	3	
L5e-B (diesel)	9	3	
L5e-A (3-wh.)	1	1	
L7e-A1 (quad)	3	3	

Dilution Tunnel vs Tailpipe

Measurements from the dilution tunnel as required by the current legislation or directly from the tailpipe can have differences in particle number concentration due to various processes like thermophoresis, agglomeration, condensation, nucleation and diffusion [20, 21]. In addition, the exhaust flow rate used in the calculations can have a big effect. The contribution of these processes to the PN results was investigated by comparing two PMP systems one measuring at the tailpipe and one at the dilution tunnel ([Figure 1](#)). The results are shown in [Figure 2](#). Initially, for some tests both PMP systems were connected to the dilution tunnel in order to confirm that they were measuring similarly. The difference was <5%. Then one of the systems was moved to the tailpipe. The mean differences ranged from -10% to +25%, with a standard deviation of 35%. It should be noted, that for the mopeds (L1e-B) the system at the tailpipe measured on average higher than the system at the dilution tunnel. The long residence time (>10 s at idle for example) in the tube between the tailpipe and the dilution tunnel probably resulted in diffusion losses and agglomeration that led to lower concentrations in the dilution tunnel. These results confirm that the results that will be presented from the dilution tunnel should be quite close to what would be measured at the tailpipe at least for solid particles >23 nm.

However, quite often mopeds and motorcycles emit particles <23 nm. In this size range the particle loss mechanisms are significant. In order to investigate the differences at sub-23 nm levels some steady state tests were conducted with the addition of a CPC with $d_{50\%}=10$ nm to the system connected to the tailpipe. The comparison of PN emissions >23 nm and >10 nm of a constant speed test can be seen in [Figure 3](#).

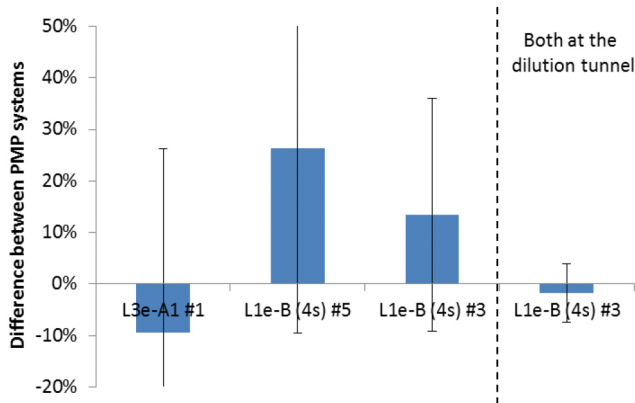


Figure 2. Difference of solid PN emissions >23 nm measured at the tailpipe compared to the full dilution tunnel system. Error bars indicate one standard deviation.

Although the agreement is very good for the >23 nm measurements (within 15%), the >10 nm measurements have differences. At the beginning of the test the system at the tailpipe is measuring almost 50% higher than the system at the dilution tunnel. The difference can be attributed to agglomeration, thermophoretic and diffusion losses. After the first 50 seconds, the PN system at the tailpipe is measuring approximately 50% lower than the system at the full dilution tunnel, which is in contrast to what would be expected. In this case the GMD (as measured at the dilution tunnel downstream of a CS with an EEPS) decreased from approximately 80 nm to 10 nm. One explanation of this unexpected behavior is that the system at the tailpipe does not count some particles because they are smaller than 10 nm. The particles when measured at the dilution tunnel are bigger due to growth from agglomeration in the tube between the tailpipe and the mixing point of the dilution tunnel. It is also possible that the condensed material on the particles is not completely removed from the PN system at the dilution tunnel; the system at the tailpipe does not have this issue because the sampling is conducted directly from the tailpipe, where the temperatures are still high and no condensation has occurred. Finally, the different penetrations of the PN systems and CPCs might have contributed, especially considering that the GMD was around 10 nm. For this test the exhaust gas temperature was less than 250 °C at the tailpipe sampling location, so no significant desorption is expected. Tests with CPCs with lower $d_{50\%}$ had even bigger differences (Figure not shown).

More detailed analysis of measurements at the dilution tunnel and the tailpipe has been conducted elsewhere [20]. There the researchers saw even bigger differences between tailpipe and dilution tunnel; the tailpipe system measuring higher. In their case the high temperatures and concentrations resulted in higher thermophoretic losses and decrease of concentration due to agglomeration. The low exhaust flowrate of the moped might also have resulted in high diffusion losses [21] due to the long residence time in the tube between the tailpipe and the dilution tunnel. This tube in many cases is corrugated and can further increase the losses. In any case, the message is clear: Particle measurements of L-category vehicles for legislative purposes need further investigation, especially for the small engines.

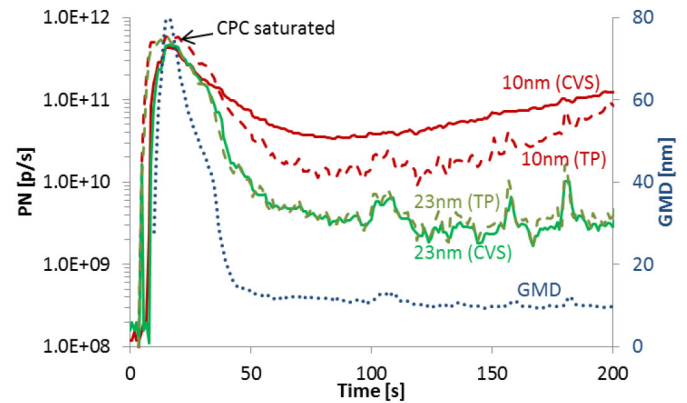


Figure 3. Comparison of measurements at the dilution tunnel (CVS) and the tailpipe (TP) (L3e-A1 #1) during a constant speed test.

Sampling Conditions

Target of these tests was to determine the minimum PCRF (dilution) for accurate measurements without interference of volatile artifact and at the same time to investigate the nature of the emitted particles. One moped and one motorcycle were tested at constant speed and the primary PCRF was changed from 15 to 1000.

4-Stroke Motorcycle

Figure 4 shows a test with a 4-stroke motorcycle at 100 km/h (L3e-A2 #3) after 5 min warm-up at 50 km/h. The EEPS was sampling directly from the dilution tunnel. The EEPS was saturated so only part of the cycle is shown. CPCs with $d_{50\%} = 23$ nm and $d_{50\%} = 3$ nm were sampling downstream of the HD+ET.

At the beginning of the 100 km/h test the GMD was 10 nm and the concentration of solid and total particles was the same. After time 400 s the concentration of total particles was higher than the solid ones, indicating formation of volatile particles.

The concentration of particles >23 nm decreased over time. At time 500 s the concentration of particles >3 nm increased to very high levels. The PCRF changed between 15×15 and 200×15 (primary × secondary PCRFs). However the concentration (corrected for the PCRF) from the different CPCs remained relatively constant indicating that these were 'solid' particles and not an artifact due to re-nucleation downstream of the ET. At this point the EEPS was connected downstream of the HD+ET and the GMD of the solid particles was around 10 nm, justifying the high difference between the 3 nm and 23 nm CPCs. These tests also show that a low PCRF (dilution) around 15×15 is appropriate for 4-stroke engines at least at warmed up operation. Nevertheless, much higher PCRF was used in this paper (>100×10).

The difference between solid particles >23 nm and >3nm is very high. The tests with high PCRF or a CS showed that they are solid (i.e. do not evaporate at 350 °C). It was the first time that the specific motorcycle was operated at this speed for so long, thus desorption of solid particles from the tubes is possible. The high exhaust gas temperature might have resulted in pyrolysis of the volatiles that were desorbed (or emitted). These sub-2.3 nm particles could also be heavy

molecular hydrocarbons that do not evaporate at 350 °C and thus are counted as solid particles. For example, in [22] the volatility analysis of particles from a 2-stroke engine showed that only 60% of the volume evaporated at a temperature of 350 °C. Finally, these particles could be metals originating from the fuel and lubricant: They partially vaporized during combustion and nucleated [23], especially in this case where the soot mode was low. This fraction could be 1% of the mass emissions [22]. Abrasion metals (e.g. Fe, Cr, Ni, Cu, Pb) from piston rings, valves and bearings are usually assumed to be low (ng/km) and in the μm range. Similarly for metals from the coatings of catalytic converters (e.g. Pt, Va). Filter analysis of exhaust gas particulate matter often finds high amount of residuals that can be ash [13, 23, 24].

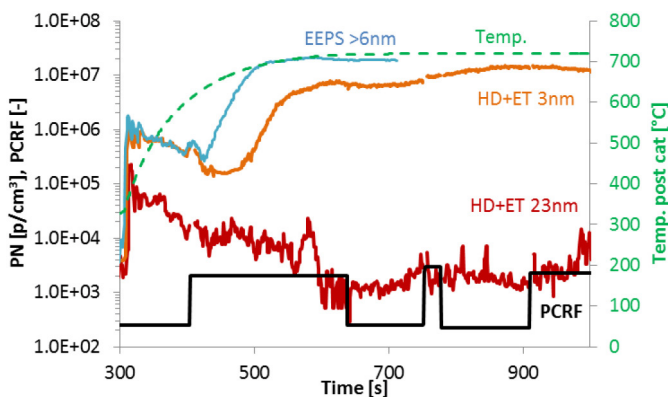


Figure 4. Investigation of PCRf effect on solid particle emissions for a 4-stroke motorcycle from the dilution tunnel (CVS) (L3e-A2 #3).

Although no re-nucleation artifacts were noticed during the hot operation of the engine in the previous example, it has to be mentioned that artifacts (i.e. re-nucleation downstream of the ET) depends on the availability of volatiles. The primary dilution affects directly the availability of volatiles, but the secondary dilution can also have an effect. The combustion process affects the absolute levels as well, thus during cold start tests and accelerations it is possible to have artifacts even with the maximum dilution. These effects were investigated with a 2-stroke moped.

2-Stroke Moped

Figure 5 shows an example of two different measurements of a 2-stroke moped (L1e-B (2s) #2). One was conducted in December 2013 (A) and the other in October 2014 (B). Although the emissions >23 nm are almost identical, the >3 nm emissions have a big difference at the beginning of the cycle. The CPC with $d_{50\%}=3$ nm was saturated during period (B) even though a 1000×10 PCRf (dilution) was used.

The nature of these particles was further investigated by using the EEPS (>6nm) at different sampling locations. Figure 6a shows the results: Sampling directly from the dilution tunnel with constant volume sampling (CVS) or the tailpipe (TP) with a catalytic stripper (CS) did not show this spike (see time around 20 s). Sampling from the CVS with a CS showed this spike which is much higher than the total particles measuring directly from the dilution tunnel (CVS). The size distributions showed a separate nucleation mode (Figure 6c). These results confirm that this high spike is re-nucleation downstream

of the ET (Figure 5) or even the CS (Figure 6). Pyrolysis inside the ET or CS cannot be excluded but is highly unlikely at the 300-350 °C temperature range.

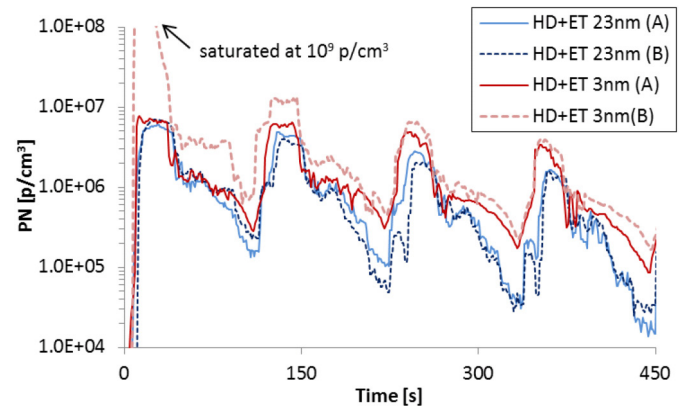


Figure 5. Investigation of artifacts for a 2-stroke moped (L1e-B (2s) #2). Measurements from the dilution tunnel (CVS).

The CS had this artifact at the dilution tunnel but not at the tailpipe. Note that the CS was used without primary but only with secondary dilution ($\times 10$) both at the tailpipe and the dilution tunnel. A possible explanation is that at the dilution tunnel the particles have grown by condensation and then the residence time inside the CS (0.1 s in this case) is not enough to completely evaporate and oxidize them.

Sampling from the tailpipe did not have this issue because the temperatures are already high. As it will be shown in the lab tests with atomized oil the concentration of hydrocarbons was at the limit of the specific CS.

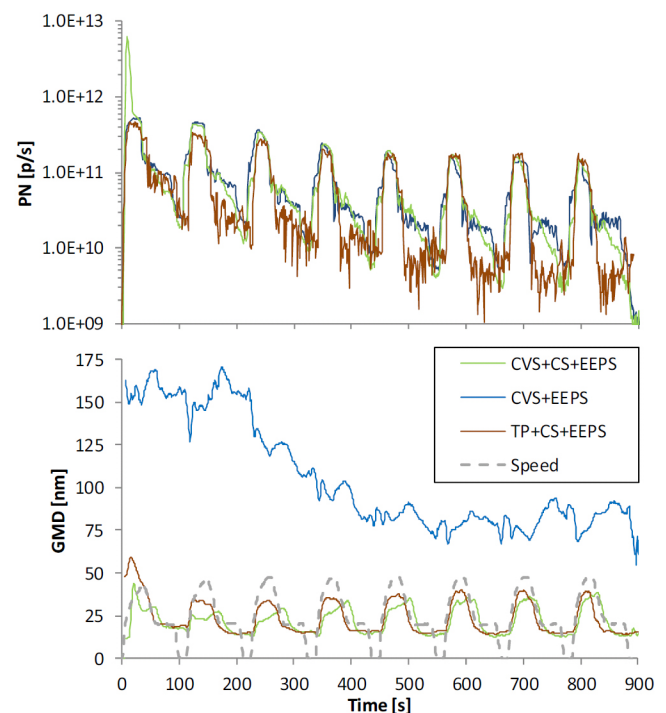


Figure 6. a. Real time emissions as measured with the EEPS with different configurations, b. GMD (average of 5 s) over time. L1e-B (2s) #2 c. example of size distributions at $t=20$ s.